Resonant Coherent Excitation of $C^{5+}$ in Si Observed with Backward Electron Spectroscopy

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Abstract

Resonant coherent excitation (RCE) of $C^{5+}$ has been observed with the loss electron yield produced by ionization of the projectiles, measured in the backward direction from a bulk Si crystal. At a resonance energy of 3.01 MeV/u for Si$(100)$, the loss electron yield obtained from the difference between the electron yield for the $C^{5+}$ and $C^{6+}$ beams has been increased by a factor of 1.2–1.3 due to RCE from the ground state to the first excited state ($n = 1$ to 2) of $C^{5+}$. The backward spectroscopy of loss electrons allows observations of RCE that is restricted within an extremely thin surface layer of the bulk crystals. In addition, the RCE-assisted electron loss process is of vital importance for precise understanding of the loss electron spectra from single crystal targets.

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1 Introduction

An axially channeled ion moving along an equally spaced row of atoms in the crystal experiences a periodic electronic field of frequency $v/d$, where $v$ is the ion velocity and $d$ is the atomic spacing. A fast ion can be excited resonantly when the frequency matches the excitation energy of the bound electron $I = Nh\nu/d$, where $h$ is the Planck constant and $N = 1, 2, 3, \ldots$ [1]. RCE thus occurs for axial and also, in a modified manner, for planar channeling. Details of RCE have been described, for example, in a review by Krause and Datz [2].

RCE has so far been observed mainly by charge-state analysis of channeled ions passed through thin, self-supported crystals [3, 4, 5]. Under such experimental conditions, RCE affects the photon emission [6], or the convoy electron emission [7]. It should be noted that RCE has been also studied using surface channeling [8, 9].

Recently, it has been demonstrated that the charge states of 2.5 and 3.5 MeV/u C ions moving along a channeling or nonchanneling direction of Si and Ge crystals are reflected in the loss electron yield which results from the ionized electrons from the projectile ions. [10]. In this case, the most probable charge state in the channeling cases is $C^{5+}$. Accordingly, we may expect to observe RCE by measurements of the loss electrons from $C^{5+}$. Under RCE conditions, the loss electron yield from $C^{5+}$ might be increased by the easier ionization from an excited state than from the ground state. This is a two-step loss process in contrast to the normal (non-resonant) loss process [11]. From a viewpoint of ion-induced electron spectroscopy, it is of essential interest to investigate the influence of RCE on the production of loss electron yield from a bulk target since, unless it is negligibly small, the resonant loss process must be taken into account when crystal targets are used.

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2 Experiments

Figure 1 schematically shows the experimental setup. The C\textsuperscript{5+} and C\textsuperscript{6+} beams in the energy range 2.8–3.2 MeV/u were obtained from the 12MV tandem accelerator at the University of Tsukuba, and were incident on a chemically cleaned Si(100) surface at room temperature under a pressure of \(\sim 8 \times 10^{-7}\) Pa. For Si(100), the RCE condition for the 2nd harmonics \((N = 2)\) from the ground state to the first excited state of C\textsuperscript{5+} is satisfied at 3.01 MeV/u \((I = 367.2\) eV, \(d = 5.43\) Å).

![Schematic illustration of the experimental setup. The first collimation slit (not shown) is placed at 1.9 m upstream of the 1-mm aperture.](Fig. 1: Schematic illustration of the experimental setup. The first collimation slit (not shown) is placed at 1.9 m upstream of the 1-mm aperture.)

The beams were collimated by double slits to an angular divergence of less than 0.015° which is much less than the channeling critical angle of Si(100), \(\sim 0.16°\). The \(\langle 100 \rangle\) axial direction perpendicular to the Si surface was chosen to observe RCE. The energy spectra of the ion-induced electrons were measured at 180° with respect to the beam direction over a solid angle of \(\sim 2 \times 10^{-3}\) sr, which is similar to that described in detail elsewhere [12]. The relative energy resolution \((\Delta E/E)\) of the spectrometer \(\sim 5\%\) is sufficient for the present analysis of the loss electron yield. Further technical aspects of the 180° electron spectroscopy combined with the ion channeling technique have been discussed in a recent literature [13].

3 Results and discussion

Figure 2 shows energy spectra of the emitted electrons induced by C\textsuperscript{5+} and C\textsuperscript{6+} at the incidence energies of 3.01 and 3.11 MeV/u under Si(100) channeling conditions. The spectra are normalized to the same number of the incident ions. The peak at \(\sim 1.5\) keV is due to Si K-shell Auger electrons. Note that the yield in the vertical axis is the number of electron signals counted. Accordingly, the spectra include no correction for the energy acceptance of the spectrometer, nor for the energy dependence of the counting efficiency of the electron multiplier. However, this does not affect the discussion below since the electron spectra are compared essentially at the same electron energy.

In Fig. 2, \(E_L\) indicates the loss-peak energy which is the kinetic energy of the electron moving at the same velocity with the ion. The ionized electrons from the ion contribute to the backward electron yield at energies lower than \(\sim E_L\). The yield below \(E_L\) for C\textsuperscript{5+} is higher than for C\textsuperscript{6+} except at energies lower than \(\sim 0.7\) keV. The higher yield results from the collision-induced loss electrons anticipated not for C\textsuperscript{6+}, but for C\textsuperscript{5+}. At the low electron energies, i.e., less than 0.7 and 0.6 for 3.01 and 3.11 MeV/u cases, respectively, the yields for C\textsuperscript{5+} are less than those for C\textsuperscript{6+} because of the reduced ionization due to the screened nuclear charge of C\textsuperscript{5+} [10].

For a better statistics, we have integrated the electron yield in a fixed energy range, for example, 1.1–1.3 keV (just below the Si K-shell Auger yield), and the integrated yields \(Y_{\text{int}}^{5+}\) and \(Y_{\text{int}}^{6+}\) for C\textsuperscript{5+} and C\textsuperscript{6+}, respectively, were used to determine the normalized loss yield \(L\), defined by,

\[
L = \frac{(Y_{\text{int}}^{5+} - Y_{\text{int}}^{6+})}{Y_{\text{int}}^{6+}}.
\]

\((1)\)

\(L\) is actually the ratio of the loss electron yield to the binary-encounter electron yield. The latter is the dominant energy-transfer process to the target electrons, as is typical for keV electron yield induced by MeV/u ions [14].
Fig. 2: Energy spectra of electrons emitted from Si induced by 3.01 (RCE condition) and 3.11 MeV/u C\(^{5+}\) and C\(^{6+}\) under Si\langle100\rangle axial incidence conditions. The yields are normalized to the same number of the incident ions. \(E_L\) indicates the loss-peak energy. The relative energy resolution of the spectrometer is \(\sim 5\%\).

Figure 3 shows the ion-energy dependence of \(\Delta\) for the 0.9–1.1, 1.1–1.3, and the wide 0.9–1.3 keV electron energy ranges. The estimated statistical error in the values of \(\Delta\) is \(\pm 4\%\). At the calculated resonance energy of 3.01 MeV/u, the enhanced value of \(\Delta\) can be clearly recognized for the three integrated ranges, which is an experimental evidence for the RCE-assisted loss electron process. While the background yields under the low-energy side of the RCE peak cannot be clearly determined, those in the high-energy side are recognizable at energies higher than \(\sim 3.05\) MeV/u. Accordingly, the RCE process enhances the loss electron yield by a factor of 1.2–1.3 at the resonance condition.

For further consideration of the present results, we refer to the published works for C\(^{5+}\) RCE of the same excitation conditions (\(I = 367.2\) eV, \(N = 2\)). Moak et al. [4] observed the RCE by charge state measurements of the ions transmitted through a Au crystal foil of 850-Å thickness. Near the RCE-matching velocity of C\(^{5+}\), the transmitted C\(^{5+}\) fraction exhibited double minima, which lies within the \(\pm 1\%\) change in the fraction, due to the Stark splitting of \(n = 2\) levels by \(\sim 4.6\) eV. To investigate such splitting, however, the present uncertainty in the \(\Delta\) values (\(\pm 4\%\)) must be much reduced, although the splitting corresponds to a resolvable C\(^{5+}\) energy of 0.075 MeV/u. In another published work, Kimura et al.[7] measured convoy electrons emitted in the forward direction of a Au crystal foil of \(\sim 1600\) Å thickness under the same RCE conditions of C\(^{5+}\). They found that the convoy yield normalized to the number of the emergent C\(^{5+}\) is enhanced by a factor of \(\sim 1.2\) near the resonance energy of 20.4 MeV. The similar amount of the enhancement factor between the loss yield (Fig. 3) and the convoy yield seems reasonable since in the latter case, also, the electron loss (to continuum) is the main production mechanism.

The resonance shape of RCE is determined by several factors such as a shift and Stark splitting of the excited energy level by the electrostatic field in the crystal, the distribution of channeled ions in the transverse space, and the thermal displacement of crystal atoms [16]. Besides these
intrinsic factors, the resonance shape can be also degraded by the energy loss of the ions in the crystal [17]. In the present case, the additional factor, i.e., the deceleration of loss electrons in the escape paths must be taken into account. Indeed, the observed shape of the RCE peak is characterized by the effective escape length of the loss electrons. Since the most probable energy of the produced loss electrons is equal to $E_L$, the typical loss yield observed at an energy of $\epsilon$ should have an escape length of $(E_L - \epsilon)/S_e$, where $S_e$ is the electron stopping power of Si. Using the value of $S_e$, for example, 1.9 and 1.6 eV/Å for 1.1 and 1.6 keV electrons, respectively [15], we obtain the mean escape length of $\sim$300 Å for the typical value of $\epsilon = 1.0$–1.2 keV. It follows that the RCE that occurs within the surface layer of $\sim$300 Å thickness is preferentially reflected in the loss electron yield. Furthermore, the energy loss of 3 MeV/u C passing through 300 Å along a Si(100) channeling direction is $\sim$18 keV, i.e., 0.0015 MeV/u, which is estimated with the assumed channeling stopping power of 60 eV/Å, i.e., 50% of the nonchanneling stopping power for Si [18]. The ratio of the ion energy loss to the ion energy at the RCE condition is 0.0015/3.01 = 0.050%. Therefore, the effect of projectile energy loss is too small to influence the observed RCE curve shown in Fig. 3.

The half width of the high-energy-side slope of the RCE peak (Fig. 3) is $\sim$0.015 MeV/u which corresponds to the resonance energy width of $367.2 \times 0.015/3.01/2 = 0.91$ eV, according to the $I \propto v$ relation (§1). On the other hand, the half width of the slope in the referred works lies in the range 4–7 eV. The sharper shoulder of the RCE peak in the present case should stem from the smaller ratio of the ion energy loss to the ion energy at the RCE condition, noted earlier. Evidently, the ratio 0.050% is a factor of less than 0.1 smaller than the ratios 0.59% for Moak et al., and 1.1% for Kimura et al.

4 Conclusion

The loss electron spectroscopy in the backward direction allows wide use of bulk crystals, rather than crystal foils, in the RCE studies. This observation technique would effectively provide RCE data for extremely thin crystals for which energy loss of the projectile ions can be completely

Fig. 3: Dependence of the normalized loss yield $L$ on the C$^ {5+}$ incidence energy, shown for the three integrated electron-energy ranges. The calculated resonance energy is 3.01 MeV/u. The solid curves are drawn to guide the eye.
neglected. Also, the present work has demonstrated that the RCE-assisted electron loss process must be taken into account for precise understanding of the loss electron spectra from single crystal targets.

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